Photodetachment study of the $1s3s4s\,^4S$ resonance in He^-

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23. Jan 1997

Abstract

A Feshbach resonance associated with the $1s3s4s\,^4S$ state of He^- has been observed in the $He(1s2s\,^3S) + e^-(\epsilon s)$ partial photodetachment cross section. The residual $He(1s2s\,^3S)$ atoms were resonantly ionized and the resulting He^+ ions were detected in the presence of a small background. A collinear laser-ion beam apparatus was used to attain both high resolution and sensitivity. We measured a resonance energy $E_r = 2.959\,255(7)$ eV and a width $\Gamma = 0.19(3)$ meV, in agreement with a recent calculation.

PACS number(s): 32.80.Fb

1 Introduction

The metastable He^- ion has received considerable attention since its discovery by Hiby in 1939 [1]. This simple three electron ion is the prototype of an unusual class of negative ions that are not stable, but rather metastable, against autodetachment. The lowest lying state is the $1s2s2p\,^4P^o$ state, which is bound by 77.51(4) meV relative to the $1s2s\,^3S$ state of He [2]. The He^- ion in this spin aligned quartet state cannot radiate and, since it is embedded in a doublet continuum, it can only autodetach via the relatively weak magnetic interactions. The varying strengths of these spin dependent interactions result in a differential metastability among the three fine structure levels. The longest lived $J = 5/2$ level has, for example, a lifetime of 350(15) μs [3]. Metastable He^- ions are therefore sufficiently long lived to pass through a typical apparatus with only minor depletion by autodetachment.

Excited states of He^-, on the other hand, decay rapidly via Coulomb autodetachment. Their presence is manifested as resonance structure in scattering cross sections close to thresholds for new chan-

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nel openings i. e. the excited state energies of the He atom. Many doublet resonances have been observed, for example, as transient intermediate states in studies of electron impact on atomic He targets [4]. Excited quartet states of He^-, however, have hitherto received far less attention. Such states appear as resonances in the photodetachment cross section and, to a lesser extent, in cross section for detachment via heavy particle collisions. The energy resolution in photodetachment measurements is typically much higher then in electron scattering experiments. This allows one to determine energies and widths of quartet states more accurately than for corresponding doublet states. Selection rules on photoexcitation from the $1s2s2p\,^4P^o$ ground state of He^- allow transitions only to ^4S, ^4P and ^4D excited states. There have been several experimental [5, 6, 7] and theoretical studies [8, 9, 10] of the photodetachment cross section of He^-. Most recently, Xi and Froese Fischer [11] have calculated the position and width of quartet states of He^- below the $He(n = 4)$ thresholds.

The prominent $1s2p^2\,^4P$ shape resonance, which lies just above the $He(1s2p\,^3P^o)$ threshold, was first observed experimentally by Peterson et al [12] in a laser photodetachment study. The accuracy of the measured resonance parameters was later improved by Walter et al [13].

In the present paper we report on the first investigation of the doubly excited $1s3s4s\,^4S$ state in He^-, which appears as a Feshbach resonance in the $He(1s2s\,^3S) + e^-(\epsilon s)$ partial cross section just below the $He(1s3s\,^3S)$ threshold.

2 Experiment

2.1 Procedure

A two-color excitation scheme (Fig. 1) was used to isolate the $He(1s2s\,^3S) + e^-(\epsilon s, \epsilon d)$ partial photodetachment cross sections in this experiment. The procedure was previously developed for studies of threshold behavior [14] and resonance structure [15] in the photodetachment cross section of Li^-. In the first step, metastable He^- ions were detached by absorption of photons of frequency ω_1. In the second step

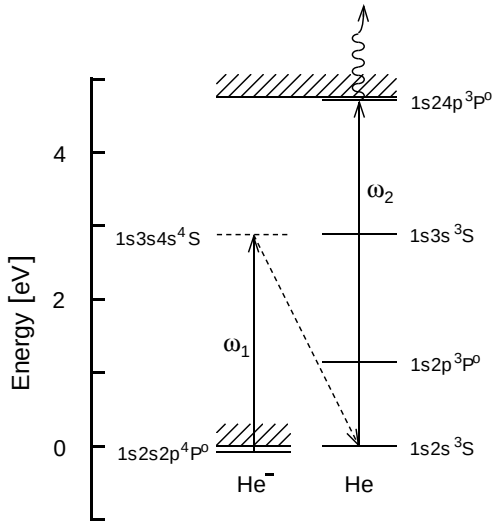

Figure 1: Excitation scheme showing selected states of He/He^-. The solid arrows represent the transitions induced in this experiment. The dashed arrow indicates one autodetachment channel of the doubly excited state of He^-. The wavy arrow indicates field ionization.

He atoms, left in the $1s2s\,^3S$ state as result of the photodetachment, were resonantly excited to a high lying Rydberg state by absorption of photons of frequency ω_2 and subsequently field ionized in a static electric field of about 150 kV/m. These processes can be described by the following equations:

$$\begin{aligned} He^-(1s2s2p\,^4P^o) + \hbar\omega_1 &\rightarrow He(1s2s\,^3S) + e^-(\epsilon s, \epsilon d), \\ He(1s2s\,^3S) + \hbar\omega_2 &\rightarrow He(1s24p\,^3P^o), \qquad (1) \\ He(1s24p\,^3P^o) &\rightsquigarrow He^+(1s\,^2S) + e^- \qquad (2) \end{aligned}$$

where $\rightsquigarrow$ represents field ionization.

The yield of the He^+ ions produced in this state selective detection scheme was recorded as a function of the frequency ω_1, while the frequency ω_2 was held constant on the transition to the Rydberg state. Thus, only photodetachment into the $He(1s2s\,^3S_1) + e^-(\epsilon s, \epsilon d)$ channels contributed to the He^+ signal. Since both laser intensities were constant during a scan, the positive ion signal, as function

of ω_1, is proportional to the $\mathrm{He}(1s2s\,^3S_1) + \mathrm{e}^-(\epsilon s, \epsilon d)$ partial photodetachment cross sections.

The detection scheme based on the selective detection of residual $\mathrm{He}(1s2s\,^3S)$ atoms was effective in eliminating a potential background source unique to measurements involving metastable negative ions. Since He^- ions are non-stable, the ion beam will contain a fraction of He atoms produced, in flight, by autodetachment. These He atoms will, however, be in the $1s^2\,^1S$ ground state and therefore will not be resonantly photoionized.

2.2 Experimental arrangement

The $^4\mathrm{He}^-$ beam was produced from a mass selected He^+ ion beam via charge exchange in a Cs vapor cell. The beam energy was 3.1 keV. A current of typically 1 nA was obtained in the interaction region.

In the interaction region shown schematically in Fig. 2, the laser and ion beams were coaxially superimposed over the 0.5 m path between the two electric quadrupole deflectors (QD1, QD2). The beam paths were defined by apertures of 3 mm diameter at both ends of the interaction region. The apparatus has been previously described in more detail [16].

The apparatus was designed to reduce the background of He^+ ions produced by double collisional detachment by installing a pair of deflection plates (DP) just before the second quadrupole deflector (QD2). The transverse electric field between the deflection plates was insufficient to field ionize the $24p$ Rydberg atoms, but strong enough to sweep collisionally created He^+ ions out of the beam.

The highly excited He atoms were field ionized by the static electric field in the second quadrupole deflector (QD2). The resulting He^+ ions were deflected into the positive ion detector (PD), where they impinged on a metal plate (MP) and produced secondary electrons that were detected with a channel electron multiplier (CEM). To monitor the He^- beam current in the Faraday cup (FC) we periodically grounded the deflection plates.

The two photon frequencies ω_1 and ω_2 used in the experiment were produced by a pair of dye lasers pumped by a common XeCl excimer laser that delivered pulses of about 15 ns duration. The visible

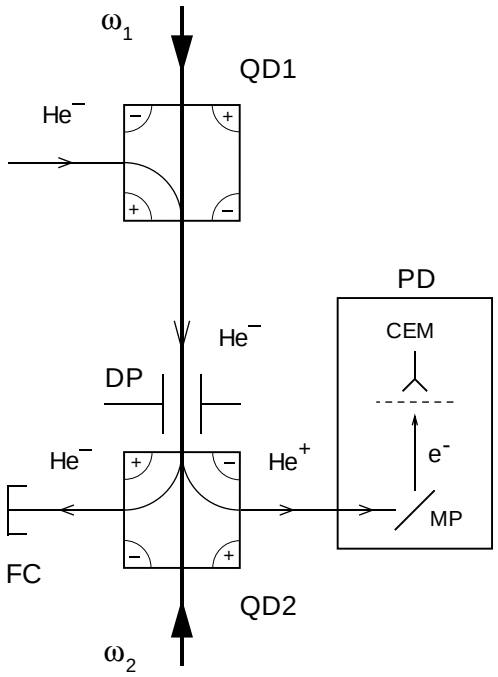

Figure 2: Interaction-detection region: QD1,QD2, electrostatic quadrupole deflectors; CEM, channel electron multiplier; DP, deflection plates; PD, positive ion detector; FC, Faraday cup; MP, metal plate. Ion- and laser beams were merged in the 0.5 m long interaction region between the quadrupole deflectors.

laser light of frequency ω_1 was generated by use of an Exalite 416 dye. The energy in this case was about 1 mJ. The UV laser light of frequency ω_2 was obtained by frequency doubling the fundamental output generated by a Coumarin 334 dye. In this case the pulse energy was typically 0.05 mJ. The output of laser ω_1 was attenuated to avoid saturation of both the photodetachment process and the detection system. The two laser pulses counter-propagated and overlapped in the interaction region.

The frequency ω_1 was determined by combining Fabry-Perot fringes with optogalvanic spectroscopy. The Fabry-Perot fringes served as frequency markers whereas the four transitions in Ar given in Table 1,

Table 1: Argon calibration lines [17]: The levels are designated in Paschen notation. For counter-propagating laser (ω_1) and ion beams we used the lines number 1, 2 and 3 and for co-propagating laser (ω_1) and ion beam we used the lines number 2, 3 and 4.

#	Transition	Wavenumber (cm^{-1})
1	$3p_5 \rightarrow 1s_4$	23 812.359(2)
2	$3p_4 \rightarrow 1s_3$	23 853.767(2)
3	$3p_8 \rightarrow 1s_5$	23 855.565(2)
4	$3p_2 \rightarrow 1s_3$	23 905.933(2)

generated in a hollow cathode lamp, provided an absolute calibration of the energy scale. The energy resolution of this apparatus has been demonstrated to be approximately 0.2 cm^{-1} [14], which is the linewidth of the laser.

3 Results and discussion

A typical measurement of the $1s3s4s\,^4S$ resonance is shown in Fig. 3. The resonance structure appears on a background created by several processes. We found three processes to significantly contribute to this background: Firstly (a), collisional detachment leaving the He atom in the $1s2s\,^3S$ state. Secondly (b), photodetachment by the laser ω_2

$$\mathrm{He}^-(1s2s2p\,^4P^{\mathrm{o}}) + \hbar\omega_2 \rightarrow \mathrm{He}(1s2s\,^3S) + \mathrm{e}^-(\epsilon s, \epsilon d),$$

followed by resonance ionization indicated in Eq. (1) and Eq. (2). Thirdly (c), non-resonant photodetachment by the laser of frequency ω_1

$$\mathrm{He}^-(1s2s2p\,^4P^{\mathrm{o}}) + \hbar\omega_1 \rightarrow \mathrm{He}(1s2s\,^3S) + \mathrm{e}^-(\epsilon d), \quad (3)$$

and then proceeding as in Eq. (1) and Eq. (2).

To reduce the collisional detached contribution (a) we maintained a pressure of 5×10^{-9} mbar (5×10^{-7} Pa) in the interaction chamber. To reduce contribution (b) we attenuated the output of laser ω_2.

The selection rule $\Delta L = 0$ for Coulomb autodetachment in LS-coupling forbids the $1s3s4s\,^4S$ resonance from appearing in the $\mathrm{He}(1s2s\,^3S) + \mathrm{e}^-(\epsilon d)$ partial cross section. Thus, the process (c), as represented by Eq. (3), contributes a constant background over the region shown in Fig. 3. Xi and Froese Fischer [18] predict a d-wave photodetachment cross section of 6 Mb across the resonance and a peak s-wave cross section of approximately 20 Mb.

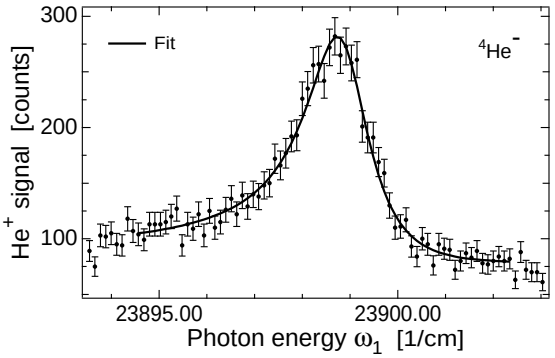

Figure 3: $\mathrm{He}^-(1s3s4s\,^4S)$ resonance: Measurement of the $\mathrm{He}(1s2s\,^3S) + \mathrm{e}^-(\epsilon s, \epsilon d)$ partial photodetachment cross sections of He^- in the vicinity of the $1s3s4s\,^4S$ resonance. The use of co-propagating laser (ω_1) and ion beams causes the resonance to be Doppler shifted by 30.89 cm^{-1} to the blue. The solid line is a fit to the data using Eq. (4) (for details, see text). The error bars represent the shot noise. Each data point represents 200 laser pulses.

The cross section $\sigma_{\mathrm{Sh}}(E)$ around the resonance can be parametrized according to Shore [19],

$$\begin{aligned} \sigma_{\mathrm{Sh}}(E) &= a + \frac{b\epsilon + c}{1 + \epsilon^2} \quad (4) \\ \epsilon &= \frac{E - E_0}{(\Gamma/2)} \quad , \end{aligned}$$

where E_0 is the resonance energy, Γ the resonance width, E the photon energy, a the background cross section, and b, c are the Shore parameters. This function is least-square fitted to our positive ion signal. The Shore parametrization method applies to either total or partial photodetachment cross sections.

The data shown in Fig. 3 are recorded with co-propagating laser (ω_1) and ion beams. The fit yields a value for the blue-shifted resonance energy E_0^{b}. To

Table 2: Comparison of the present measurement and a calculation of the $1s3s4s\,^4S$ resonance in He^-. The recommended conversion factor of (1/8 065.5410) $\left[\mathrm{eV}/(\mathrm{cm}^{-1})\right]$ was used [20].

Author	E_r (eV)	Γ (meV)
Theory:		
Xi et al [11] (1996)		
Length form	2.959 07	0.19
Velocity form	2.959 08	0.18
Experiment:		
This work	2.959 255(7)	0.19(3)

eliminate the Doppler shift, we repeated the measurements using counter-propagating laser and ion beams to determine the red-shifted resonance energy, E_0^r. The resonance energy, corrected for the Doppler effect to all orders, E_r, is given by the geometric mean of the two measurements:

$$\begin{aligned} E_r &= \sqrt{E_0^b E_0^r} \\ &= 23\,867.992(55)\ \mathrm{cm}^{-1} \quad . \end{aligned}$$

This result is an average obtained from 7 spectra taken with co-propagating laser and ion beams and 8 spectra taken with counter-propagating beams. There are two major contributions to the quoted uncertainty: 0.01 cm^{-1} is due to the calibration uncertainty, and the remainder is due to statistical scatter of the fitted resonance parameters. In Table 2 the values are compared with recently calculated resonance parameters.

The measured resonance parameters agree with those calculated by Xi and Froese Fischer [11] within the limited precision of the latter. Our measurement is, however, almost two orders of magnitudes more precise and it should stimulate further theoretical work.

4 Summary

In the present experiment we have combined laser photodetachment and resonance ionization to study a Feshbach resonance associated with the autodetaching decay of the double excited $1s3s4s\,^4S$ state in He^-. The measurement was made under the simultaneous conditions of high sensitivity and energy resolution using a collinear beam apparatus. The production of doubly excited states was enhanced by the use of a large interaction volume defined by the overlap of the co-axially superposed ion and laser beams. The residual atoms produced in the photodetachment process were selectively detected according to their excitation state to reduce the background. The energy resolution is enhanced in a collinear beam measurement by kinematic compression of the longitudinal velocity distribution of the fast moving ions [21]. In addition, the Doppler shift was removed, to *all* orders, by performing separate measurements using co- and counter-propagating laser and ion beams.

The frequency doubled output of excimer pumped dye lasers can be used, in principle, to study the photodetachment cross section of He^- up to the double detachment limit. Moreover, the decay of a given doubly excited state can be studied in different channels. We plan to search for resonances below higher lying thresholds and to investigate different decay channels.

5 Acknowledgement

Xi and Froese Fischer are acknowledged for having provided us with unpublished data. Financial support for this research has been obtained from the Swedish Natural Science Research Council (NFR). Personal support was received from Wenner-Gren Center Foundation for Igor Kiyan and from Nordisk Forskerutdanningsakademi (NorFA) for James Peterson. David Pegg acknowledges the support from the Swedish Institute and the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences.

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